

PAT-NO: JP405238830A

DOCUMENT-IDENTIFIER: JP 05238830 A

TITLE: **SINTERED ALUMINUM NITRIDE AND ITS PRODUCTION**

PUBN-DATE: September 17, 1993

INVENTOR-INFORMATION:

NAME

KOMATSU, MICHIMASU

NAKAMURA, MIHO

ASSIGNEE-INFORMATION:

NAME

TOSHIBA CORP

COUNTRY

N/A

APPL-NO: JP04035245

APPL-DATE: February 21, 1992

INT-CL (IPC): C04B035/58

ABSTRACT:

PURPOSE: To obtain **sintered aluminum nitride** having high strength, thermal conductivity and heat-radiation performance by heating a formed material composed of **AlN** powder and a grain boundary forming component under prescribed condition and solidifying the product by cooling at a prescribed cooling rate.

CONSTITUTION: A **sintered AlN** having high strength and thermal conductivity and a **grain boundary size** and maximum **pore** diameter of $\leq 1\mu\text{m}$ can be produced by adding 1-7.5wt.% of a grain boundary forming component (rare earth element and/or alkaline earth metal element) to **AlN** powder, forming and degreasing the obtained raw material mixture, **sintering** at $1700-2000^\circ\text{C}$ and solidifying the formed liquid phase formed from the grain boundary component by cooling the **sintered** compact at a cooling rate of $<100^\circ\text{C/h}$ until the temperature is lowered to the freezing temperature of the liquid phase.

COPYRIGHT: (C)1993,JPO&Japio

BEST AVAILABLE COPY

(19)日本国特許庁 (J P)

(12) 公開特許公報 (A)

(11)特許出願公開番号

特開平5-238830

(43)公開日 平成5年(1993)9月17日

(51)Int.Cl.⁵

C 0 4 B 35/58

識別記号

I 0 4 B 8821-4G

F 8821-4G

庁内整理番号

F I

技術表示箇所

審査請求 未請求 請求項の数 4 (全 7 頁)

(21)出願番号 特願平4-35245

(22)出願日 平成4年(1992)2月21日

(71)出願人 000003078

株式会社東芝

神奈川県川崎市幸区堀川町72番地

(72)発明者 小松 通泰

神奈川県横浜市鶴見区末広町2の4 株式

会社東芝京浜事業所内

(72)発明者 中村 美保

神奈川県横浜市鶴見区末広町2の4 株式

会社東芝京浜事業所内

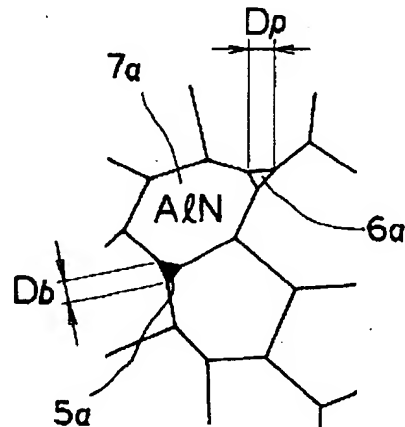
(74)代理人 弁理士 波多野 久 (外1名)

(54)【発明の名称】 窒化アルミニウム焼結体およびその製造方法

(57)【要約】

【目的】本発明の目的は、高強度で熱伝導率が高く放熱特性が優れた窒化アルミニウム焼結体およびその製造方法を提供することにある。

【構成】本発明に係る窒化アルミニウム焼結体の製造方法は、窒化アルミニウムに粉末に対して、粒界相形成成分として希土類元素およびアルカリ土類金属元素の少なくとも一方を添加した原料混合体を成形脱脂し、得られた成形体を1700~2000℃の焼結温度で所定時間加熱焼結した後に、上記焼結温度から、上記希土類元素および/またはアルカリ土類金属元素により焼結時に形成された液相が凝固する温度までに至る焼結体の冷却速度を毎時100℃以下に設定したことを特徴とする。



1

【特許請求の範囲】

【請求項1】 窒化アルミニウム結晶組織に希土類元素およびアルカリ土類金属の少なくとも一方を含む粒界相が形成され、粒界相の最大径が $1\mu\text{m}$ 以下であることを特徴とする窒化アルミニウム焼結体。

【請求項2】 窒化アルミニウム結晶組織に介在する気孔の最大径が $1\mu\text{m}$ 以下であることを特徴とする請求項1記載の窒化アルミニウム焼結体。

【請求項3】 粒界相を構成する希土類元素およびアルカリ土類金属の少なくとも一方の焼結体に対する含有量が1～7.5重量%であることを特徴とする請求項1記載の窒化アルミニウム焼結体。

【請求項4】 窒化アルミニウムに粉末に対して、粒界相形成成分として希土類元素およびアルカリ土類金属元素の少なくとも一方を添加した原料混合体を成形脱脂し、得られた成形体を $1700\sim 2000^{\circ}\text{C}$ の焼結温度で所定時間加熱焼結した後に、上記焼結温度から、上記希土類元素および／またはアルカリ土類金属元素により焼結時に形成された液相が凝固する温度までに至る焼結体の冷却速度を毎時 100°C 以下に設定したことを特徴とする窒化アルミニウム焼結体の製造方法。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は窒化アルミニウム焼結体およびその製造方法に係り、高強度で熱伝導率が高く放熱特性に優れた窒化アルミニウム焼結体およびその製造方法に関する。

【0002】

【従来の技術】従来の金属材料と比較して強度、耐熱性、耐食性、耐摩耗性、軽量性などの諸特性に優れたセラミックス焼結体が、半導体、電子機器材料、エンジン用部材、高速切削工具用材料、ノズル、ベアリングなど、従来の金属材料の及ばない苛酷な温度、応力、摩耗条件下で使用される機械部品、構造物や装飾品材料として広く利用されている。

【0003】特に窒化アルミニウム(A1N)焼結体は高熱伝導性を有する絶縁体であり、シリコン(Si)に近い熱膨張係数を有することから高集積化した半導体装置の放熱板や基板として、その用途を拡大している。

【0004】従来、上記窒化アルミニウム焼結体は一般的に下記の製造方法によって量産されている。すなわち、セラミックス原料としての窒化アルミニウム粉末に焼結助剤と、有機バインダと、必要に応じて各種添加剤や溶媒、分散剤とを添加して原料混合体を調製し、得られた原料混合体をロール成形法やドクターブレード法によって成形し、薄板状ないしシート状の成形体として、原料混合体をプレス成形して厚板状ないし大型の成形体を形成する。次に得られた成形体は、空気または窒素ガス雰囲気において $400\sim 500^{\circ}\text{C}$ に加熱され脱脂処理され、有機バインダとして添加された炭化水素成分

2

等が成形体から排除脱脂される。そして脱脂された成形体は窒素ガス雰囲気等で高温に加熱され緻密化焼結されて窒化アルミニウム焼結体が形成される。

【0005】上記焼結操作は、一般に図3に示すように、例えば黒鉛(グラファイト)製の炉材を貼設した焼成炉1の炉床2上に箱状の焼成容器3を配置し、この焼成容器3内に1個または複数の脱脂した窒化アルミニウム成形体4を収容した状態で加熱し、所定の焼結温度 $1700\sim 2000^{\circ}\text{C}$ に4～8時間保持した後に、焼成炉1の加熱用電源をOFFにして焼結体を炉冷して実施していた。

【0006】上記窒化アルミニウム成形体4を収容保持する焼成容器3や炉床2は、高温焼結時に成形体と反応して焼結体の特性を低下させることを防止するために、また不純物による焼結体の汚染を防止するために、成形体と同一材料である高純度の窒化アルミニウム(A1N)焼結体や窒化硼素(BN)焼結体で形成される。

【0007】上記のように窒化アルミニウム成形体4と反応せず熱容量が大きい焼成容器中に成形体4を収容して焼結しているため、各窒化アルミニウム成形体4の全体に熱が均一に作用し、むらの少ない均質な焼結体得られる。また焼成炉1の炉材に含まれる炭素等の不純物が、焼成容器に遮断され焼成時に直接的に成形体4に作用することがなく、これらの不純物によって焼結体に色むらや変形が発生したり、焼結体の熱伝導性が低下することが防止される。上記焼成容器3を使用せず、窒化アルミニウム成形体4を直接焼成炉1内に配置して焼成した場合には、焼成炉1の炉材から放出された過剰量の炭素蒸気や不純物によって焼結体表面が著しく損傷され、かつ焼結体全体の変形量も大きくなり、製品歩留りが急減してしまう。

【0008】上記製造方法において、原料A1N粉末として平均粒径が $0.3\mu\text{m}$ 以下程度の超微細な原料粉末を使用する場合は、A1N粉末単独でもかなりの緻密な焼結体得られる。しかしながら、原料粉末表面等に付着した多量の酸素等の不純物が焼結時に、A1N結晶格子中に固溶したり、格子振動の伝播を妨げるA1-O-N化合物等の複合酸化物を生成する結果、焼結助剤を使用しないA1N焼結体の熱伝導率は比較的に低かった。

【0009】一方原料粉末として平均粒径 $0.5\mu\text{m}$ 以上のA1N粉末を使用する場合は、その原料粉末単独では焼結性が良好でないため、ホットプレス法以外には助剤無添加では緻密な焼結体を得ることが困難であり、量産性が低い欠点があった。そこで常圧焼結法によって効率的に焼結体を量産しようとする場合には、焼結体の緻密化およびA1N原料粉末中の不純物酸素がA1N結晶粒子内へ固溶することを防止するために、焼結助剤として、酸化イットリウム(Y_2O_3)などの希土類酸化物や酸化カルシウムなどのアルカリ土類金属酸化物等を添加することが一般に行なわれている。

【0010】これらの焼結助剤は、AlN原料粉末に含まれる不純物酸素と反応してイットリウム-アルミニウム-ガーネット (YAG, $3Y_2O_3 \cdot 5Al_2O_3$)、イットリア-アルミナ化合物 (YAL, $Y_2O_3 \cdot Al_2O_3$)、イットリア-アルミナ-金属化合物 (YAM, $2Y_2O_3 \cdot Al_2O_3$) などから成る液相を形成し、焼結体の緻密化を達成するとともに、この不純物酸素を粒界相として固定し、高熱伝導率化も達成するものと考えられている。またこれらの液相は焼結後においてAlN結晶粒の粒界部にガラス質または結晶質として凝固して粒界相を形成し、この粒界相がAlN結晶粒を相互に強固に結合せしめAlN焼結体全体の強度を高めると考えられている。

【0011】

【発明が解決しようとする課題】しかしながら従来の製造方法においては、原料粉末の平均粒径、焼結助剤の種類および添加量、脱脂焼結条件等を厳正に管理した場合においても、焼結体の強度が不足して製品歩留りが低下したり、所定の熱伝導率が得られず、AlNの最大利用特性である優れた放熱特性が損われる場合が多かった。

【0012】本発明は上記問題点を解決するためになされたものであり、高強度で熱伝導率が高く放熱特性が優れた窒化アルミニウム焼結体およびその製造方法を提供することを目的とする。

【0013】

【課題を解決するための手段】本願発明者らは上記目的を達成するため、まず焼結体の強度や熱伝導率が低下する原因を実験により解明することを試みた。すなわち原料窒化アルミニウム粉末に添加する焼結助剤や添加物の種類および添加量、不純物の残留量、成形焼結条件、焼結体の組成等を種々変えて、それらが焼結体特性に及ぼす影響や関係について実験検討を進め、以下に示すように知見を得た。

【0014】すなわち本発明者らは加熱焼結操作完了直後における焼結体の冷却速度の大小が、最終的に製造される焼結体の品質特性に大きな影響を及ぼすことを突き止めた。

【0015】すなわち、従来の製造方法においては、成形体を所定の焼結温度で一定時間加熱保持して緻密化焼結を実施した後に、焼成炉の加熱用電源をOFFとし焼結体を炉冷していたため、焼成炉の形式によって差はあるが、焼結体の冷却速度が毎時400~800℃程度と極めて大きな値となっており、焼結体の窒化アルミニウム結晶組織が粗雑になる大きな原因であることが確認された。X線回折図や走査型電子顕微鏡写真等で焼結体の破面を観察したところ、図2に示すように、直径Dbが5~10μm程度の粗大な酸化物粒界相5や直径Dpが50~100μm程度の粗大な気孔6がAlN結晶粒7の粒界部に多数形成されていることが判明した。これらの粒界相5は、焼結助剤として添加した Y_2O_3 などの

希土類化合物が焼結時に窒化アルミニウム原料粉末表面のアルミニウム酸化物と反応して生成した液相(成分: YAG, YAL, YAMなど)が冷却時に凝集偏析して形成されたものである。また粗大な粒界相5の周辺には液相がなくなった気孔6が形成され、上記粒界相5および気孔6は共に窒化アルミニウム焼結体の熱伝導を妨げる抵抗として作用するとともに、気孔6は未焼結部となりAlN結晶粒7相互の接合強度が低下し、焼結体全体としての強度が低下してしまうことが判明した。

10 【0016】一方、焼成炉の加熱装置に対する通電量を制御して焼結直後の焼結体の冷却速度を、従来の炉冷による冷却速度より低く設定して得られた焼結体の結晶組織を観察し、また焼結体の各種特性を測定した。その結果、図1に示すようにいずれも窒化アルミニウム結晶組織の粒界相5aの直径Dbが小さく、液相の凝集偏析がなく、微細な粒界相が均一に分布した結晶組織が得られた。また気孔6aについても直径Dpが小さく、凝集が少ない均一分布を有する組織が得られ、高い熱伝導率および高強度を備えるAlN焼結体を得られた。

20 【0017】本発明は上記知見に基づいて完成されたものである。すなわち本発明に係るセラミックス焼結体の製造方法は、窒化アルミニウムに粉末に対して、粒界相形成成分として希土類元素およびアルカリ土類金属元素の少なくとも一方を添加した原料混合体を成形脱脂し、得られた成形体を1700~2000℃の焼結温度で所定時間加熱焼結した後に、上記焼結温度から、上記希土類元素および/またはアルカリ土類金属元素により焼結時に形成された液相が凝固する温度までに至る焼結体の冷却速度を毎時100℃以下に設定したことを特徴とする。

30 【0018】さらに本発明に係る窒化アルミニウム焼結体は、窒化アルミニウム結晶組織に希土類元素およびアルカリ土類金属の少なくとも一方を含む粒界相が形成され、粒界相の最大径が1μm以下であることを特徴とする。

【0019】また窒化アルミニウム結晶組織に介在する気孔の最大径は1μm以下に設定するとよい。

【0020】さらに粒界相を構成する希土類元素およびアルカリ土類金属の少なくとも一方の焼結体に対する含有量は1~7.5重量%に設定するとよい。

【0021】本発明方法において使用され、焼結体の主成分となる窒化アルミニウム(AlN)粉末としては、焼結性および熱伝導性を考慮して不純物酸素含有量が7重量%以下、好ましくは3重量%以下に抑制され平均粒径が0.05~5μm程度、好ましくは3μm以下のものを使用する。

【0022】希土類元素およびアルカリ土類金属は焼結助剤として窒化アルミニウム原料粉末に添加される。焼結助剤の具体例としては希土類元素(Y, La, Sc, Pr, Ce, Nd, Dy, Gdなど)の酸化物、窒化

5

物、アルカリ土類金属 (Ca, Ba, Sr など) の酸化物、もしくは焼結操作によりこれらの化合物となる物質 (炭酸塩等) が単独で、または2種以上混合して使用され、特に酸化イットリウム (Y_2O_3) や酸化カルシウム (CaO) が好ましい。これらの焼結助剤は、窒化アルミニウムの原料粉末表面のアルミニウム酸化物相と反応して複合酸化物 ($Al_5Y_3O_{12}$, $AlYO_3$, $Al_2Y_4O_9$ など) の液相を形成し、この液相が焼結体の高密度化 (緻密化) をもたらす。例えば Y_2O_3 を焼結助剤として用いた場合、アルミン酸イットリウムが生成し液相焼結が進行すると考えられる。これらの焼結助剤を添加して常圧焼結すると、焼結性の向上 (緻密化) のみではなく、熱伝導率も向上できる。すなわち焼結時に AlN 中に固溶していた不純物酸素が Y_2O_3 と反応して結晶粒界の酸化物相として偏析するため、格子欠陥の少ない焼結体を得られ、熱伝導率が向上する。

【0023】上記焼結助剤の添加量は1~7.5重量%の範囲で調整される。添加量が1重量%未満の場合は、焼結性の改善効果が充分に発揮されず、焼結体が緻密化されず低強度の焼結体が形成されたり、 AlN 結晶中に酸素が固溶し、高い熱伝導率を有する焼結体が形成できない。一方添加量が7.5wt%を超える過量となると、過量の粒界相が焼結体中に残存したり、熱処理により除去される粒界相の体積が大きいため、焼結体中に空隙 (気孔) が残ったりして収縮率が増大し、変形を生じ易くなる。

【0024】窒化アルミニウム結晶組織に形成される粒界相および気孔の大きさは焼結体の伝熱特性および強度特性に大きく影響するため、本発明に係る焼結体においては粒界相の最大径は1 μm 以下に設定される一方、気孔の最大径は1 μm 以下に設定される。粒界相の最大径が1 μm を超えるように液相の凝集偏析が著しくなると、 AlN 結晶粒子相互の液相による結合作用が低下し焼結体全体としての強度が低下し易くなると同時に粗大な粒界相は熱伝導の妨げとなり、焼結体の熱伝導率を低下させる。

【0025】一方、気孔についても同様にその最大径が1 μm を超えるように粗大となると伝熱抵抗が高くなり焼結体の熱伝導率が低下するとともに焼結体の強度低下が著しくなる。

【0026】上記のように窒化アルミニウム結晶組織に形成される粒界相の最大径を1 μm 以下に、また気孔の最大径を1 μm 以下にするためには、焼結操作完了直後における焼結体の冷却速度を毎時100℃以下調節することが必要である。上記冷却速度を毎時100℃を超えるように設定した場合には、焼結体に生成した液相が粒界部に凝集偏析し易く、粗大な粒界相および気孔が形成されてしまう。

【0027】上記冷却速度を調節する温度範囲は、所定の焼結温度 (1700~2000℃) から、前記の焼結

6

助剤の反応によって生じる液相が凝固するまでの温度 (液相凝固点) までで充分である。前記のような焼結助剤を使用した場合の液相凝固点は概略1650~1500℃程度である。こうして少なくとも焼結温度から液相凝固点に至るまでの焼結体の冷却速度を毎時100℃以下に制御することにより、微細な粒界相が AlN 結晶粒周囲に均一に分布し、気孔の形成が少ない焼結体を得られる。

【0028】次に上記窒化アルミニウム焼結体を製造する場合の概略工程について説明する。すなわち窒化アルミニウムに所定量の焼結助剤、有機バインダ等の必要な添加剤を加えて原料混合体を調製し、次に得られた原料混合体を成形して所定形状の成形体を得る。原料混合体の成形法としては、汎用の金型プレス法、静水圧プレス法、あるいはドクターブレード法、ロール成形法のようなシート成形法などが適用できる。

【0029】上記成形操作に引き続いて、成形体を非酸化性雰囲気中、例えば窒素ガス雰囲気中で温度400~500℃で1~2時間加熱して、予め添加していた有機バインダを充分に除去する。

【0030】次に脱脂処理された成形体は、焼成容器内に収容して焼成炉内において多段に積層され、この配置状態で複数の成形体は一括して所定温度で焼結される。焼結操作は、窒素ガスなどの非酸化性雰囲気中で成形体を温度1700~2000℃に2~10時間程度加熱して実施される。焼結雰囲気は、窒素ガス、または窒素ガスを含む還元性雰囲気で行なう。還元性ガスとしては H_2 ガス、 CO ガスを使用してもよい。なお、焼結は真空 (僅かな還元雰囲気を含む)、減圧、加圧および常圧を含む雰囲気で行なってもよい。焼結温度が1750℃未満と低温状態で焼成すると、原料粉末の粒径、含有酸素量によって異なるが、緻密な焼結体を得にくい一方、2000℃より高温で焼成すると、焼成炉内における AlN 自体の蒸気圧が高くなり緻密化が困難になるおそれがあるため、焼結温度は上記範囲に設定される。

【0031】上記焼結操作において緻密な焼結体を得るためにも、また焼結体の熱伝導率を向上させるためにも、ある程度の焼結助剤の添加は必要である。しかしながら、焼結助剤は AlN や不純物酸素と反応して $Al_5Y_3O_{12}$, $AlYO_3$, $Al_2Y_4O_9$ などの酸化物を形成して粒界相に析出する。これら粒界相の酸化物は熱伝導を妨げる作用を有することが確認されている。したがって過剰量の粒界相が形成されないように焼結助剤の添加量は厳正に管理する必要がある。

【0032】上記製法によって製造された窒化アルミニウム焼結体は、いずれも多結晶体として非常に高い200w/m·k (25℃) に近い熱伝導率を有し、また曲げ強度等の機械的特性にも優れている。

【0033】

【作用】上記構成に係る窒化アルミニウム焼結体および

その製造方法によれば、焼結処理完了直後における焼結体の冷却速度を毎時100℃以下と小さく設定しているため、炉冷のような急速冷却を実施した場合と異なり、焼結時に生成した液相の凝集偏析が少なく、微細な粒界相が均一に分布した結晶組織が得られる。また結晶組織に形成される気孔も小形化すると同時に減少させることができる。したがって、粗大な粒界相や気孔によって熱伝達や緻密化が阻害されることが少なく、高強度で高い熱伝導率を有する窒化アルミニウム焼結体を得られる。

【0034】

【実施例】次に下記の実施例を参照して本発明に係る窒化アルミニウム焼結体およびその製造方法による効果をより具体的に説明する。

【0035】実施例1～3

不純物として酸素を1.0重量%含有し、平均粒径1.5μmの窒化アルミニウム粉末に対して、焼結助剤としてのY₂O₃（酸化イットリウム）を5重量%添加し、エチルアルコール中で30時間湿式混合した後に乾燥して原料粉末混合体を調製した。次に乾燥して得た原料粉末混合体をプレス成形機の成形用金型内に充填して1200kg/cm²の加圧力にて圧縮成形して円板状放熱板の成形体を多数調製し、引き続き各成形体を空気中で温度375℃で2時間加熱して脱脂処理した。

【0036】次に前記工程で脱脂処理した複数の成形体を、図3に示すように2個ずつまとめて高純度A1N製焼成容器3内に収容し、この4個の焼成容器3をN₂ガスを封入した焼成炉内に2段に積層配置した。そして焼成炉1内の温度を1815℃まで高めた状態で4時間保持し、緻密化焼結を実施した後に、焼成炉に付設した加

熱装置への通電量を減少させて焼成炉内温度が1500℃まで降下するまでの間における焼結体の冷却速度がそれぞれ100℃/hr（実施例1）、50℃/hr（実施例2）、25℃/hr（実施例3）、となるように調整して焼結体を冷却した。その結果、それぞれ直径100mm、厚さ3.0mmである実施例1～3に係るA1Nセラミックス焼結体を8個ずつ調製した。

【0037】比較例1

一方、緻密化焼結完了直後に、加熱装置電源をOFFにし、従来の炉冷による冷却速度（約500℃/hr）で焼結体を冷却した点以外は実施例1と同一条件で焼結処理して同一寸法を有する比較例1に係るA1N焼結体を調製した。

【0038】比較例2

また、緻密化焼結完了直後における焼結体の冷却速度を250℃/hrと過大に設定した以外は実施例1と同一条件で焼結処理して同一寸法を有する比較例2に係るA1N焼結体を調製した。

【0039】そして得られた実施例1～3および比較例1～2に係る各窒化アルミニウム焼結体の特性を評価するため、各焼結体表面部をX線解析法によって分析し、さらに各焼結体の破面を走査型電子顕微鏡によって観察することによって、粒界相の最大径、気孔の最大径を測定するとともに、窒化アルミニウム結晶組織における粒界相の凝集の有無および気孔の凝集の有無を観察した。さらに各焼結体の熱伝導率および曲げ強度の平均値を測定し、下記表1右欄に示す結果を得た。

【0040】

【表1】

試料 No	焼結後の 1500℃までの 冷却速度 (℃/Hr)	粒界相の 最大径 (μm)	粒界相の 凝集の有無	気孔の 最大径 (μm)	気孔の 凝集の有無	熱伝導率 ($\text{W}/\text{m} \cdot \text{K}$)	曲げ強度 (kg/mm^2)
実施例 1	100	< 1	無	< 1	無	188	40
実施例 2	50	< 1	無	< 1	無	195	45
実施例 3	25	< 1	無	< 1	無	197	48
比較例 1	500	< 15	有	< 5	有	155	30
比較例 2	250	< 5	有	< 2	有	170	36

【0041】表1に示す結果から明らかなように、実施例1～3に係る窒化アルミニウム焼結体においては、比較例1～2と比較して緻密化焼結完了直後における焼結体の冷却速度を従来法より低く設定しているため、結晶組織内において液相の凝集偏析が少なく、また気孔の凝集もなかった。顕微鏡観察したところ、結晶組織はいずれも図1に示すように粒界相5aの最大径Dbが1 μm 未満と小さく、また気孔6aの最大径Dpも1 μm 未満と微小であった。そして微細な粒界相が均一に分布した結晶組織であるため、高密度（高強度）で高熱伝導度を*50

*有する放熱性の高い焼結体を得られた。

【0042】一方、比較例1および2のように焼結体の冷却速度を大きく設定し、急激に冷却した場合には、図2に示すように最大径Dbが15 μm となるような粗大な粒界相5が形成さ、また最大径Dpが4 μm と大きな気孔6が各所に観察され、焼結性が低下して強度および熱伝導率も低下した。

【0043】

【発明の効果】以上説明の通り本発明に係る窒化アルミニウム焼結体およびその製造方法によれば、焼結処理完

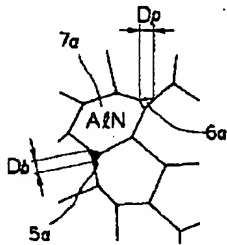
11

了直後における焼結体の冷却速度を毎時100℃以下と小さく設定しているため、炉冷のような急速冷却を実施した場合と異なり、焼結時に生成した液相の凝集偏析が少なく、微細な粒界相が均一に分布した結晶組織が得られる。また結晶組織に形成される気孔も小形化すると同時に減少させることができる。したがって、粗大な粒界相や気孔によって熱伝達や緻密化が阻害されることが少なく、高強度で高い熱伝導率を有する窒化アルミニウム焼結体が得られる。

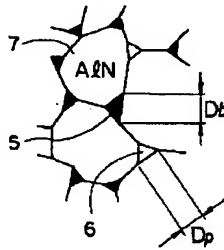
【図面の簡単な説明】

【図1】本発明に係る窒化アルミニウム焼結体の結晶組織を模式的に示す図。

【図1】



【図2】



12

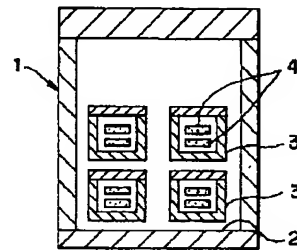
【図2】従来の窒化アルミニウム焼結体の結晶組織を模式的に示す図。

【図3】焼成容器内に複数の成形体を収容し同時に焼成する状態を示す焼成炉の断面図。

【符号の説明】

- 1 焼成炉
- 2 炉床
- 3 焼成容器
- 4 成形体
- 10 5, 5a 粒界相
- 6, 6a 空孔（気孔）
- 7, 7a AlN結晶粒

【図3】



* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] The alumimium nitride sintered compact which the grain boundary phase containing either [at least] rare earth elements or alkaline earth metal is formed in the alumimium nitride crystalline structure, and is characterized by the overall diameter of a grain boundary phase being 1 micrometer or less.

[Claim 2] The alumimium nitride sintered compact according to claim 1 characterized by the overall diameter of the pore by which it is placed between the alumimium nitride crystalline structures being 1 micrometer or less.

[Claim 3] The alumimium nitride sintered compact according to claim 1 characterized by the content to one [at least] sintered compact of the rare earth elements which constitute a grain boundary phase, and alkaline earth metal being 1 - 7.5 % of the weight.

[Claim 4] Shaping cleaning of the mixture is carried out. the raw material which added either [at least] rare earth elements or an alkaline-earth-metal element as a grain boundary phase formation component to powder to alumimium nitride -- After carrying out predetermined time heating sintering of the acquired Plastic solid at the sintering temperature of 1700-2000 degrees C The manufacture approach of the alumimium nitride sintered compact characterized by setting the cooling rate of the sintered compact which results by the temperature which the liquid phase formed at the time of sintering with the above-mentioned rare earth elements and/or an alkaline-earth-metal element solidifies from the above-mentioned sintering temperature as 100 degrees C [or less]/h.

[Translation done.]

* NOTICES *

JPO and INPIT are not responsible for any damages caused by the use of this translation.

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to an alumimium nitride sintered compact and its manufacture approach, and relates to the alumimium nitride sintered compact thermal conductivity excelled [sintered compact] in the heat dissipation property highly with high intensity, and its manufacture approach.

[0002]

[Description of the Prior Art] the ceramic sintered compact which be excellent in many properties , such as reinforcement , thermal resistance , corrosion resistance , abrasion resistance , and lightweight nature , as compared with the conventional metallic material be widely use as the cruel temperature which the conventional metallic material do not attain to , stress and the machine parts use under wear conditions , such as a semi-conductor , an electronic equipment ingredient , a member for engines , a charge of high speed cutting tool lumber , a nozzle , and a bearing , and a structure material and an accessories ingredient .

[0003] Especially an alumimium nitride (AlN) sintered compact is an insulator which has high temperature conductivity, and the application is expanded as the heat sink and substrate of the semiconductor device integrated highly from having a coefficient of thermal expansion near silicon (Si).

[0004] Conventionally, generally the above-mentioned alumimium nitride sintered compact is mass-produced by the following manufacture approach. namely, the alumimium nitride powder as a ceramic raw material -- sintering acid, an organic binder, and the need -- responding -- various additives, and a solvent and a dispersant -- adding -- a raw material -- the raw material which prepared the mixture and was obtained -- a mixture -- roll diffusion bonding and a doctor blade method -- fabricating -- considering as the Plastic solid of the shape of the shape of sheet metal, and a sheet **** -- a raw material -- press forming of the mixture is carried out and the shape of a thick plate and a large-scale Plastic solid are formed. Next, in air or nitrogen-gas-atmosphere mind, the acquired Plastic solid is heated by 400-500 degrees C, cleaning processing is carried out, and abatement cleaning of the hydrocarbon component added as an organic binder is carried out from a Plastic solid. And by nitrogen-gas-atmosphere mind etc., the degreased Plastic solid is heated by high temperature, eburnation sintering is carried out, and an alumimium nitride sintered compact is formed.

[0005] The above-mentioned sintering actuation arranges the box-like baking container 3 on the hearth 2 of the firing furnace 1 which stuck the internal insulation made from a graphite (graphite), as generally shown in drawing 3 . After having heated where alumimium nitride Plastic solid 4 which one piece or plurality degreased is held in this baking container 3, and holding in predetermined sintering temperature of 1700-2000 degrees C for 4 to 8 hours, the power source for heating of a firing furnace 1 was turned OFF, furnace cooling of the sintered compact was carried out, and it was carried out.

[0006] the above -- alumimium nitride -- a Plastic solid -- four -- hold -- maintenance -- carrying out -- baking -- a container -- three -- a hearth -- two -- an elevated temperature -- sintering -- the time -- a Plastic solid -- reacting -- a sintered compact -- a property -- falling -- making -- things -- preventing -- a sake -- moreover -- an impurity -- depending -- a sintered compact -- contamination -- preventing -- a sake -- a Plastic solid -- the same -- an ingredient -- it is -- a high grade -- alumimium nitride (AlN) -- a sintered compact -- boron nitride -- (-- BN --) -- a sintered compact -- forming -- having .

[0007] It does not react with aluminum nitride Plastic solid 4 as mentioned above, but since heat capacity has held and sintered Plastic solid 4 in a large baking container, heat acts on each whole aluminum nitride Plastic solid 4 at homogeneity, and a homogeneous sintered compact with little unevenness is obtained. Moreover, impurities, such as carbon contained in the internal insulation of a firing furnace 1, are intercepted by the baking container, and do not act on Plastic solid 4 directly at the time of baking, an irregular color and deformation occur in a sintered compact with these impurities, or it is prevented that the thermal conductivity of a sintered compact falls. The above-mentioned baking container 3 will not be used, a sintered compact front face will be remarkably damaged with the carbon steam and impurity of an excessive amount which were emitted from the internal insulation of a firing furnace 1 when aluminum nitride Plastic solid 4 was arranged and calcinated in the direct firing furnace 1, and the deformation of the whole sintered compact will also become large, and the product yield will decrease rapidly.

[0008] In the above-mentioned manufacture approach, when using the overly detailed raw material powder whose mean particle diameter is 0.3-micrometer or less extent as raw material AlN powder, a precise sintered compact also with a remarkable AlN powder independent is obtained. However, as a result of impurities', such as a lot of oxygen's adhering to a raw material powder front face etc., dissolving in an AlN crystal lattice at the time of sintering or generating multiple oxides, such as an aluminum-O-N compound which bars propagation of lattice vibration, the thermal conductivity of the AlN sintered compact which does not use sintering acid was low in comparison.

[0009] the case where AlN powder with a mean particle diameter of 0.5 micrometers or more is used as raw material powder on the other hand -- the raw material powder -- if independent, since the degree of sintering was not good, in addition to hot pressing, it is difficult to obtain a sintered compact precise assistant additive-free, and there was a fault with low mass production nature. Then, when it is going to mass-produce a sintered compact efficiently with an ordinary pressure sintering process, in order to prevent that the eburnation of a sintered compact and the impurity oxygen in AlN raw material powder dissolve into an AlN crystal grain child, generally adding alkaline-earth-metal oxides, such as rare earth oxides, such as oxidation ITTOURIMU (Y₂ O₃), and a calcium oxide, etc. is performed as sintering acid.

[0010] These sintering acid reacts with the impurity oxygen contained in AlN raw material powder. An yttrium aluminum garnet (YAG, and 3Y₂ O₃ and 5aluminum 2O₃), While forming the liquid phase which consists of a yttria-alumina compound (YAL, and Y₂ O₃ and aluminum 2O₃), yttria-alumina-metallic compounds (YAM, and 2Y₂ O₃ and aluminum 2O₃), etc. and attaining the eburnation of a sintered compact This impurity oxygen is fixed as a grain boundary phase, and it is thought that high temperature conductivity-ization is also attained. Moreover, it is thought that it solidifies in the grain boundary section of AlN crystal grain as glassiness or a crystalline substance, a grain boundary phase is formed in it, this grain boundary phase makes it combine AlN crystal grain mutually firmly, and these liquid phase raises the reinforcement of the whole AlN sintered compact to it after sintering.

[0011]

[Problem(s) to be Solved by the Invention] However, in the conventional manufacture approach, the reinforcement of a sintered compact was insufficient, when the mean particle diameter of raw material powder, the class of sintering acid and an addition, cleaning sintering conditions, etc. were managed strictly, the product yield did not fall, or predetermined thermal conductivity was not obtained, but the outstanding heat dissipation property which is the maximum utilization property of AlN was spoiled in many cases.

[0012] It is made in order that this invention may solve the above-mentioned trouble, and it aims at offering the aluminum nitride sintered compact thermal conductivity is high and the heat dissipation property excelled [sintered compact] in high intensity, and its manufacture approach.

[0013]

[Means for Solving the Problem] Invention-in-this-application persons tried to solve the cause that the reinforcement and the thermal conductivity of a sintered compact fall first, by experiment in order to attain the above-mentioned object. That is, various presentations of the class of the sintering acid added to raw material aluminum nitride powder or additive and an addition, the amount of residuals of an impurity, shaping sintering conditions, and a sintered compact etc. were changed, and experiment examination was advanced about the effect and relation which they exert on a sintered compact property, and knowledge was acquired as shown below.

[0014] That is, this invention persons traced that the size of the cooling rate of the sintered compact immediately after the completion of heating sintering actuation had big effect on the quality characteristic of the sintered compact manufactured eventually.

[0015] That is, in the conventional manufacture approach, although there be a difference according to the format of a firing furnace since the power source for heating of a firing furnace be set to OFF and furnace cooling of the sintered compact be carried out after carry out fixed time amount heating maintenance of the Plastic solid at predetermined sintering temperature and carry out eburnation sintering, it be checked that it be the big cause by which the cooling rate of a sintered compact serve as about per hour 400-800 degrees C and a very big value, and the alumimium nitride crystalline structure of a sintered compact become coarse. It is a diameter Db as it is shown in drawing 2, when the fracture surface of a sintered compact is observed with X diffraction drawing, a scanning electron microscope photograph, etc. The big and rough oxide grain boundary phase 5 and big and rough diameter Dp which are about 5-10 micrometers It became clear that much big and rough pores 6 which are about 50-100 micrometers were formed in the grain boundary section of the AlN crystal grain 7. Y2 O3 which added these grain boundary phases 5 as sintering acid etc. -- at the time of cooling, the liquid phase (component: YAG, YAL, YAM, etc.) which the rare earth compound generated in response to the time of sintering with the aluminum oxide of an alumimium nitride raw material powder front face carries out condensation segregation, and is formed. Moreover, it became clear that the pore 6 whose liquid phase was lost is formed around the big and rough grain boundary phase 5, pore 6 serves as the non-sintered section while both the above-mentioned grain boundary phase 5 and the pore 6 act as resistance which bars heat conduction of an alumimium nitride sintered compact, the bonding strength between AlN crystal grain 7 fell, and the reinforcement as the whole sintered compact fell.

[0016] On the other hand, the amount of energization to the heating apparatus of a firing furnace was controlled, and it gazed at the crystalline structure of the sintered compact which set up lower than the cooling rate by the conventional furnace cooling the cooling rate of the sintered compact immediately after sintering, and was obtained, and the various properties of a sintered compact were measured.

Consequently, as shown in drawing 1, all are the diameters Db of grain boundary phase 5a of the alumimium nitride crystalline structure. It is small, there is no condensation segregation of the liquid phase, and the crystalline structure from which the detailed grain boundary phase was distributed over homogeneity was obtained. Moreover, it is a diameter Dp also about pore 6a. It was small, the organization for which condensation has little homogeneity distribution was obtained, and the AlN sintered compact equipped with high thermal conductivity and high intensity was obtained.

[0017] This invention is completed based on the above-mentioned knowledge. Namely, the manufacture approach of the ceramic sintered compact concerning this invention Shaping cleaning of the mixture is carried out. the raw material which added either [at least] rare earth elements or an alkaline-earth-metal element as a grain boundary phase formation component to powder to alumimium nitride -- After carrying out predetermined time heating sintering of the acquired Plastic solid at the sintering temperature of 1700-2000 degrees C It is characterized by setting the cooling rate of the sintered compact which results by the temperature which the liquid phase formed at the time of sintering with the above-mentioned rare earth elements and/or an alkaline-earth-metal element solidifies from the above-mentioned sintering temperature as 100 degrees C [or less]/h.

[0018] The grain boundary phase containing either [at least] rare earth elements or alkaline earth metal is formed in the alumimium nitride crystalline structure, and the alumimium nitride sintered compact furthermore applied to this invention is characterized by the overall diameter of a grain boundary phase being 1 micrometer or less.

[0019] Moreover, the overall diameter of the pore by which it is placed between the alumimium nitride crystalline structures is good to set it as 1 micrometer or less.

[0020] The content to one [at least] sintered compact of the rare earth elements which furthermore constitute a grain boundary phase, and alkaline earth metal is good to set up to 1 - 7.5% of the weight.

[0021] It is used in this invention approach, and as alumimium nitride (AlN) powder used as the principal component of a sintered compact, an impurity oxygen content is preferably controlled to 3 or less % of the weight 7 or less % of the weight in consideration of a degree of sintering and thermal conductivity, and mean particle diameter uses preferably about 0.05-5 micrometers of things 3 micrometers or less.

[0022] Rare earth elements and alkaline earth metal are added by alumimium nitride raw material powder as sintering acid. As an example of sintering acid, the oxide of rare earth elements (Y, La, Sc, Pr, Ce, Nd, Dy, Gd, etc.), a nitride, the oxide of alkaline earth metal (calcium, Ba, Sr, etc.), or the matter (carbonate etc.) that serve as these compounds by sintering actuation are independent, or it is used by two or more sorts, mixing, and especially yttrium oxide (Y_2O_3) and a calcium oxide (CaO) are desirable. These sintering acid reacts with the aluminum oxide phase of the raw material powder front face of alumimium nitride, the liquid phase of multiple oxides (aluminum $5Y_3O_{12}$, $AlYO_3$, aluminum $2Y_4O_9$, etc.) is formed, and this liquid phase brings about the densification (eburnation) of a sintered compact. For example, Y_2O_3 When it uses as sintering acid, an ulmin acid yttrium generates and it is thought that liquid phase sintering advances. If these sintering acid is added and ordinary pressure sintering is carried out, not only the improvement in a degree of sintering (eburnation) but thermal conductivity can improve. That is, the impurity oxygen which was dissolving in AlN at the time of sintering is Y_2O_3 . In order to react and to segregate as an oxide phase of the grain boundary, a sintered compact with few lattice defects is obtained, and thermal conductivity improves.

[0023] The addition of the above-mentioned sintering acid is adjusted in 1 - 7.5% of the weight of the range. When an addition is less than 1 % of the weight, the improvement effect of a degree of sintering is not fully demonstrated, eburnation of the sintered compact is not carried out, the sintered compact of low strength is formed, or oxygen dissolves during an AlN crystal, and the sintered compact which has high thermal conductivity cannot be formed. On the other hand, if an addition turns into an excess amount exceeding 7.5wt(s)%, the grain boundary phase of an excess amount remains in a sintered compact, or since the volume of the grain boundary phase removed by heat treatment is large, a hole (pore) will remain into a sintered compact, contraction will increase, and it will become easy to produce deformation.

[0024] Since the grain boundary phase formed in the alumimium nitride crystalline structure and the magnitude of pore influence greatly the heat-conducting characteristic and strength property of a sintered compact, while the overall diameter of a grain boundary phase is set as 1 micrometer or less in the sintered compact concerning this invention, the overall diameter of pore is set as 1 micrometer or less. If the condensation segregation of the liquid phase becomes remarkable so that the overall diameter of a grain boundary phase may exceed 1 micrometer, a big and rough grain boundary phase will serve as hindrance of heat conduction, and will reduce the thermal conductivity of a sintered compact at the same time the binding action by the liquid phase between AlN crystal grain children falls and it becomes easy for the reinforcement as the whole sintered compact to fall.

[0025] If it becomes big and rough so that the overall diameter may exceed 1 micrometer similarly about pore, while thermal resistance will become high and the thermal conductivity of a sintered compact will fall on the other hand, lowering of a sintered compact on the strength becomes remarkable.

[0026] In order to set to 1 micrometer or less the overall diameter of the grain boundary phase formed in the alumimium nitride crystalline structure as mentioned above and to set the overall diameter of pore to 1 micrometer or less, it is required to adjust 100 degrees C [or less]/h of cooling rates of the sintered compact immediately after the completion of sintering actuation. When the above-mentioned cooling rate is set up so that it may exceed 100 degrees C/h, it will be easy to carry out condensation segregation of the liquid phase generated to the sintered compact at the grain boundary section, and a big and rough grain boundary phase and pore will be formed.

[0027] Even temperature (liquid phase congealing point) until the liquid phase produced by the reaction of the aforementioned sintering acid congeals from predetermined sintering temperature (1700-2000 degrees C) is enough as the temperature requirement which adjusts the above-mentioned cooling rate. The liquid phase congealing point at the time of using the above sintering acid is about 1650-1500 degrees C of outlines. In this way, by controlling the cooling rate of a sintered compact until it results [from sintering temperature] in the liquid phase congealing point at least at 100 degrees C [or less]/h, a detailed grain boundary phase is distributed over the perimeter of AlN crystal grain at homogeneity, and a sintered compact with little formation of pore is obtained.

[0028] Next, the outline process in the case of manufacturing the above-mentioned alumimium nitride sintered compact is explained. namely, alumimium nitride -- required additives, such as sintering acid of the specified quantity, and an organic binder, -- adding -- a raw material -- the raw material which prepared the

mixture and then was obtained -- a mixture is fabricated and the Plastic solid of a predetermined configuration is acquired. a raw material -- as a method of fabricating a mixture, a general-purpose die-press method, the hydrostatic-pressure pressing method or a doctor blade method, a sheet forming method like roll diffusion bonding, etc. are applicable.

[0029] To the above-mentioned shaping actuation, succeedingly, a Plastic solid is heated at the temperature of 400-500 degrees C in a non-oxidizing atmosphere, for example, nitrogen-gas-atmosphere mind, for 1 to 2 hours, and the organic binder added beforehand is fully removed.

[0030] Next, the Plastic solid by which cleaning processing was carried out is held in a baking container, a laminating is carried out into a firing furnace multistage, and two or more Plastic solids in this arrangement condition are put in block, and are sintered at predetermined temperature. By non-oxidizing atmospheres, such as nitrogen gas, sintering actuation heats a Plastic solid in temperature of 1700-2000 degrees C for about 2 to 10 hours, and is carried out. A sintered atmosphere is performed by the reducing atmosphere containing nitrogen gas or nitrogen gas. As reducibility gas, it is H₂. Gas and CO gas may be used. In addition, sintering may be performed in the ambient atmosphere containing a vacuum (few reducing atmosphere is included), reduced pressure, application of pressure, and ordinary pressure. Since there is a possibility that the vapor pressure of the AlN in a firing furnace itself may become high, and eburation may become difficult when it calcinates by high temperature from 2000 degrees C while it will be hard to obtain a precise sintered compact although it changes with the particle size of raw material powder, and amounts of content oxygen if sintering temperature calcinates in the state of less than 1750 degrees C and low temperature, sintering temperature is set as the above-mentioned range.

[0031] Also in order to obtain a precise sintered compact in the above-mentioned sintering actuation, and also in order to raise the thermal conductivity of a sintered compact, a certain amount of sintering acid needs to be added. however, sintering acid -- AlN and impurity oxygen -- reacting -- aluminum 5Y3 -- O₁₂, AlYO₃, and aluminum 2Y₄O₉ etc. -- an oxide is formed and it deposits in a grain boundary phase. It is checked that the oxide of these grain boundary phase has the operation which bars heat conduction. Therefore, it is necessary to manage the addition of sintering acid strictly so that the grain boundary phase of an excessive amount may not be formed.

[0032] Each alumimium nitride sintered compact manufactured by the above-mentioned process has the thermal conductivity near 200 w/m-k (25 degrees C) very high as the polycrystalline substance, and is excellent also in mechanical properties, such as flexural strength.

[0033]

[Function] According to the alumimium nitride sintered compact concerning the above-mentioned configuration, and its manufacture approach, since the cooling rate of the sintered compact immediately after the completion of sintering processing is small set up with 100 degrees C [or less]/h, unlike the case where forced cooling like furnace cooling is carried out, there are few condensation segregations of the liquid phase generated at the time of sintering, and the crystalline structure from which the detailed grain boundary phase was distributed over homogeneity is obtained. Moreover, it can be made to decrease while the pore formed in the crystalline structure is also miniaturized. Therefore, it is rare for heat transfer and eburation to be checked by a big and rough grain boundary phase and pore, and the alumimium nitride sintered compact which has high thermal conductivity with high intensity is obtained.

[0034]

[Example] Next, the effectiveness by the alumimium nitride sintered compact applied to this invention with reference to the following example and its manufacture approach is explained more concretely.

[0035] after containing oxygen 1.0% of the weight as one to example 3 impurity, adding Y₂O₃ (oxidization yttrium) as sintering acid 5% of the weight to alumimium nitride powder with a mean particle diameter of 1.5 micrometers and carrying out wet blending in ethyl alcohol for 30 hours -- drying -- raw material powder -- the mixture was prepared. next, the raw material powder dried and obtained -- a mixture -- the inside of the molding die of a press-forming machine -- being filled up -- 1200kg/cm² It pressed with welding pressure and many Plastic solids of a disc-like heat sink were prepared, each Plastic solid was succeedingly heated at the temperature of 375 degrees C in air for 2 hours, and cleaning processing was carried out.

[0036] Next, as shown in drawing 3 , it holds at a time collectively two or more two Plastic solids which carried out cleaning processing at said process in the baking container 3 made from a high grade AlN, and it

is N2 about these four baking containers 3. Laminating arrangement was carried out in two steps into the firing furnace which enclosed gas. And the temperature in a firing furnace 1 is held in the condition of having raised to 1815 degrees C for 4 hours. The cooling rate of the sintered compact which can be set by the time it decreases the amount of energization to the heating apparatus attached to the firing furnace and whenever [firing furnace internal temperature] descends to 1500 degrees C, after carrying out eburnation sintering, respectively 100 degrees C/hr (example 1), It adjusted and the sintered compact was cooled so that it might be set to 50 degrees C/hr (example 2) and 25 degrees C/hr (example 3). Consequently, it prepared at a time eight AlN ceramic sintered compacts concerning the examples 1-3 which are 3.0mm in the diameter of 100mm, and thickness, respectively.

[0037] The heating apparatus power source was turned OFF immediately after example of comparison 1 one side, and the completion of eburnation sintering, and the AlN sintered compact concerning the example 1 of a comparison which carries out sintering processing on the same conditions as an example 1, and has the same dimension was prepared except the point which cooled the sintered compact with the cooling rate (about 500 degrees C/hr) by the conventional furnace cooling.

[0038] The AlN sintered compact concerning the example 2 of a comparison which carries out sintering processing on the same conditions as an example 1, and has the same dimension was prepared except having set the cooling rate of the example 2 of a comparison, and the sintered compact immediately after the completion of eburnation sintering as 250 degrees C/hr and fault size.

[0039] And in order to evaluate the property of each aluminum nitride sintered compact concerning the examples 1-3 and the examples 1-2 of a comparison which were acquired, while measuring the overall diameter of a grain boundary phase, and the overall diameter of pore by analyzing each sintered compact surface section by the X-ray **** method, and observing the fracture surface of each sintered compact with a scanning electron microscope further, the existence of condensation of the grain boundary phase in the aluminum nitride crystalline structure and the existence of condensation of pore were observed. Furthermore the thermal conductivity of each sintered compact and the average of flexural strength were measured, and the result shown in the following table 1 right column was obtained.

[0040]

[A table 1]

試料 No.	焼結後の 1500℃までの 冷却速度 (℃/Hr)	粒界相の 最大径 (μm)	粒界相の 凝集の有無	気孔の 最大径 (μm)	気孔の 凝集の有無	熱伝導率 ($\text{W/m}\cdot\text{K}$)	曲げ強度 (kg/mm^2)
実施例 1	100	< 1	無	< 1	無	188	40
実施例 2	50	< 1	無	< 1	無	195	45
実施例 3	25	< 1	無	< 1	無	197	48
比較例 1	500	< 15	有	< 5	有	155	30
比較例 2	250	< 5	有	< 2	有	170	36

[0041] Since it compared with the examples 1-2 of a comparison and the cooling rate of the sintered compact immediately after the completion of eburnation sintering was set up in the alumimium nitride sintered compact concerning examples 1-3 lower than a conventional method so that clearly from the result shown in a table 1, into the crystalline structure, there were few condensation segregations of the liquid phase, and condensation of pore did not have them, either. When microscope observation is carried out, as each crystalline structure shows drawing 1, it is the overall diameter Db of grain boundary phase 5a. It is small and is [less than 1 micrometer and] the overall diameter Dp of pore 6a. It was as minute as less than

1 micrometer. And since a detailed grain boundary phase was the crystalline structure distributed over homogeneity, the high sintered compact of the heat dissipation nature which has high temperature conductivity by high density (high intensity) was obtained.

[0042] when the cooling rate of a sintered compact is greatly set up like the examples 1 and 2 of a comparison on the other hand and it cools rapidly, it is shown in drawing 2 -- as -- overall diameter Db the big and rough grain boundary phase 5 which is set to 15 micrometers -- formation -- moreover, overall diameter Dp 4 micrometers and the big pore 6 were observed by every place, the degree of sintering fell, and reinforcement and thermal conductivity also fell.

[0043]

[Effect of the Invention] According to the alumimium nitride sintered compact applied to this invention above as explanation, and its manufacture approach, since the cooling rate of the sintered compact immediately after the completion of sintering processing is small set up with 100 degrees C [or less]/h, unlike the case where forced cooling like furnace cooling is carried out, there are few condensation segregations of the liquid phase generated at the time of sintering, and the crystalline structure from which the detailed grain boundary phase was distributed over homogeneity is obtained. Moreover, it can be made to decrease while the pore formed in the crystalline structure is also miniaturized. Therefore, it is rare for heat transfer and eburnation to be checked by a big and rough grain boundary phase and pore, and the alumimium nitride sintered compact which has high thermal conductivity with high intensity is obtained.

[Translation done.]

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.